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A Review Of Plutonium (Pu) Combustion Releases In Air For Inhalation Hazard Evaluation

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A Review Of Plutonium (Pu) Combustion Releases In Air For Inhalation Hazard Evaluation*

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ABSTRACT

Experimental data are compiled and reviewed for aerosol particle releases due to combustion in air of Plutonium (Pu). The aerosol release fraction (ARF), which is the mass of Pu aerosolized, divided by the mass of Pu oxidized, is dependent on whether the oxidizing Pu sample is static (i.e. stationary) or dynamic (i.e. falling in air). ARF data are compiled for sample masses ranging from 30 mg to 1770 g, oxidizing temperatures varying from 113 °C to ~1000 °C, and air flow rates varying from 0.05 m/s to 5.25 m/s. The measured ARFs range over five orders of magnitude. The maximum observed static ARF is 2.4×10^{-3}, and this is the recommended ARF for safety studies of static Pu combustion.

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INTRODUCTION AND MOTIVATION

The Annular Core Research Reactor (ACRR) in Technical Area 5 is a pool-type research reactor capable of pulsed, steady-state, and transient-rod-withdrawal operations. Experiments with plutonium (Pu) are being planned for the ACRR and regulations require an evaluation of the hypothetical airborne releases from such experiments. The purpose of this report is to review and evaluate previous experiments in which the aerosolized plutonium release fraction from combustion was measured, and to provide guidance on how these measurements may be used to assess a hypothetical inhalation hazard in the ACRR.

The two critical coefficients needed to evaluate an aerosol hazard are the airborne or aerosol release fraction (ARF) and the respirable fraction (RF). The ARF is defined as the mass fraction of the affected sample that is aerosolized by the process of interest. The particles that form the aerosol range in size from nanometers to tens or even hundreds of micrometers in diameter. Because solid particles in nature are often irregular in shape, a standardized diameter called the aerodynamic diameter is used to evaluate and compare aerosol particles with different shapes and material densities. The aerodynamic diameter of a particle is defined as the diameter of a perfect sphere of material with a density of 1 gram/cm^3 (sometimes called unit density), that has the same settling velocity as the particle of interest. The fraction of inhaled particles that deposit in the respiratory tract, (and may therefore have serious health consequences), is primarily a function of the particle aerodynamic diameter. This respirable deposition fraction will, however, vary from person to person and is also dependent on the activity level of the person inhaling

the aerosol. A representative plot of the respirable deposition fraction as a function of the aerodynamic diameter, averaged over variations in the population, is shown in Figure 1 [Hinds, 1999]. In Figure 1 the total respirable fraction deposited in the respiratory tract is shown as a solid line. This total fraction is made up of contributions by deposition in three regions; the head airways, the tracheobronchial passageways, and the lung alveoli. The fractions deposited in these regions are shown separately as broken lines. Particles deposited in the lower parts of the respiratory tract (i.e. the tracheobronchial and alveolar regions) are not readily expelled from the body and are called respirable particles. From Figure 1 we see that such particles need to be less than 10 to 20 μm diameter aerodynamic. Therefore, the respirable fraction (RF) of an aerosol can be defined as the mass fraction of aerosol particles with aerodynamic diameters smaller than 10 μm [DOE Handbook 3010-94, 1994, page 1-4].

In the experiments reviewed in this work, the reported aerosol mass is for particles with aerodynamic diameters that are less than or equal to 10 μm. This implies that the RF is one, which is the highest and therefore most conservative RF associated with the reported ARFs.

An excellent compilation of the available data on the ARF and RF for Pu combustion is summarized in the Department of Energy (DOE) Handbook 3010-94 [1994]. However, to avoid misapplication, handbook values should not be used without fully understanding the origin of the numbers. It is because of possible misquotations [Haschke and Martz, 1998], omissions, and misunderstandings that may occur from using a compilation, that

the original literature for Pu oxidation in air will be again compiled and evaluated in this report.

LITERATURE REVIEW

The ARF is defined as the mass of Pu that is aerosolized and entrained in the air stream, divided by the mass of oxidized Pu. Thus, unless the entire Pu sample is oxidized, the ARF will be larger than the aerosolized Pu mass divided by the Pu sample mass. In this work, oxidation is used to describe the process in which Pu is exposed to air and releases particles containing Pu, regardless of the chemical composition of the Pu-bearing aerosol particles. Thus, reactions between Pu and water vapor may also be occurring that forms Pu-bearing aerosol particles.

The experimental data base for determining the aerosol release fraction (ARF) for oxidizing pieces of Pu can be divided into essentially two categories based on the conditions of the experiment. In the first category, the sample is supported and is stationary. In these so-called static tests, there is no bulk motion of the sample during oxidation. In the second category the sample is unsupported and is falling in air. For these dynamic tests, Pu is oxidized while falling and may continue oxidizing after impacting a surface.

The ARF (aerosol release fraction) data for static oxidation are summarized in Table I and in the first two columns of Table II. The data in Table I are also displayed in Figures 2, 3, and 4. The static test data that are labeled “All Static Oxidation Results” in Figure 5

are tabulated in the first two columns of Table II. Dynamic oxidation data are given in the last two columns of Table II and displayed in Figure 5 with the label "All 0.75 m Fall Results". The static test data given in Table I and Figures 2, 3, and 4 are in the mass range of 1.95 g to 1770 g, and the details of the test conditions associated with sample mass are given in Table I. The dynamic test ARF data in Table II and Figure 5 are for sample masses in the range of 30 mg to 2.5 g, however the specific ARF for a given sample mass is unavailable.

The original source for the ARF data in Figure 5 is from Carter and Stewart [1971], as given in their Figure 13. The available copy of Figure 13 by Carter and Stewart is difficult to read and was therefore digitized and redrawn below as Figure 5. (Two sets of the digitized data are also given in Table II.) The legend, axis scaling, and axis labels in Figure 5 are taken from Figure 13 of Carter and Stewart. Although not stated in the original work, the plot was probably generated by first arranging the ARF measurements in increasing order. If a total of N measurements were performed for a given data set, then the i-th data point would correspond to an x-axis value of (100)(i)/N, and the measured ARF would be plotted on the y-axis. (Because there is no point plotted for 100% on the x-axis, this last data point may have been excluded.) The cumulative probability x-axis scale used in their work is such that the data fall on a straight line if the logarithm of the fraction of airborne aerosol less than 10 μm aerodynamic is normally distributed. The mean value for any of the four test conditions is given for an x-axis value of 50%. Because of inaccuracies in digitizing an already optically digitized figure,

the values given in Figure 5 are probably accurate to within only ±5%. The data for all the static and dynamic tests are given as circles and diamonds, respectively.

Static Oxidation Experiments

All the experiments given in Table I are for static experiments that were performed in the 1960's. Table entries for reports in the 1990's are either rejected for the reasons stated or are reanalysis of the original experiments. The experiments seemed to be mostly scoping tests to determine what if any aerosols are produced, and to provide some measure of the releases. In many cases, repeating tests with identical conditions were generally not performed to obtain good statistics. Instead the experiments were often conducted by varying several parameters such as sample mass, sample geometry, Pu phase, sample holding device, temperature, air humidity, and airflow configuration. Thus it is difficult to discern trends in the data or determine the critical variables. Furthermore, although today aerosol instruments are available to provide real time measurements of aerosol particle size-resolved concentrations, no such instruments were available in the 1960's. Thus the data are generally time integrations of the release fractions. Nonetheless, given the safety constraints and problems of conducting Pu combustion tests in a glove box, the data are very valuable and represent the state-of-the-art at the time the tests were conducted.

As shown in Figure 2, the data may be grouped into four sets; (1) oxidation tests below the ignition temperature, labeled "Stewart 1-9", (2) oxidation tests at or above the

ignition temperature, labeled “Stewart 10-15”, (3) oxidation tests inside a furnace, labeled “Mishima 1-6”, and (4) oxidation test for Pu ignited with a torch, labeled “Mishima 7-9”. This grouping into four sets of tests is also used in Figures 3 and 4.

Stewart [1963] performed the first two sets of tests. These tests are listed in Table I as tests S1 to S9 and S10 to S15, respectively, and are shown as unfilled squares or circles, respectively in Figures 2, 3, and 4. In these tests, samples were suspended in the airflow and oxidized by heat from a furnace. The sample temperature, air humidity, Pu phase, and sample mass were varied. The ARFs vary from 5.6×10^{-7} to 2.4×10^{-3}.

Mishima [1965, 1966] performed the third and fourth sets of tests. The data are listed in Table I as tests M1 to M6 and M7 to M9, respectively, and are shown as filled squares and circles in Figures 2, 3, and 4, respectively. For tests M1 to M6, samples were oxidized in a furnace, and the sample temperature and air flow rate varied. However, the sample size and geometry were essentially constant. The samples were suspended in a quartz boat that partially obstructed the airflow around the sample. The ARFs vary from 2.8×10^{-8} to 5.3×10^{-7}. For the fourth set of tests, samples were resting on a thermally insulating flat surface and ignited by a torch. The sample size, geometry, and Pu phase were varied and the ARFs vary from 3.4×10^{-5} to 4.7×10^{-4}.

The static tests performed by Carter and Stewart [1971] consisted of burning metal fragments without sample movement. The samples were suspended at the top of a 0.75 m long and 0.14 m diameter column, and heated with a resistance furnace. The residue

crumbled, disintegrated on cooling, and fell into the column efficiently dispersing the fine particles. The specimens were delta phase, alloyed metal as thin rod or foil, with surface areas to mass ratios of 2 to 4 cm^2/g. Sample masses ranged from 30 mg to 2.5 g, but most were in the range 40 to 200 mg. Unfortunately, details of each test such as the initial mass or oxidation temperature corresponding to a measured ARF are not available. Therefore, these data are plotted separately in Figure 5 and listed separately in Table II.

Analysis of Static Experiments

It is difficult to discern a clear trend in the ARF data from Figures 2, 3 or 4. Sample mass does not seem to have a clear influence on the ARF. From Figure 2 we see that although the Mishima tests (M1-M6) and those of Stewart (S1-S15) are for comparable sample masses, the ARFs are orders of magnitude lower than those for Stewart. Unfortunately, these two investigators did not perform their tests under similar conditions so comparisons are difficult. In Figures 3 and 4 the same data are plotted based on the sample temperature or airflow velocity, respectively. If the Mishima experiments (M1-M6) are ignored in Figure 3, then there is some tendency for the ARF to increase with temperature.

In reviewing all the data in Table I for static oxidation, even with airflow velocities up to 5.25 m/s, the maximum reported ARF is 2.4×10^{-3}. This high ARF reported for experiment S12 in which "the temperature of the specimen fluctuated markedly and was in the region of the melting point for only a fraction of the time. The relatively large

fraction airborne during these experiments was due probably to the contractions and expansions accompanying the temperature changes." [Stewart, 1963]. This description supports the idea that oxide crust formation and spallation leads to high aerosol releases, and not merely turbulent airflow.

The lack of apparent trends in the data has been noted in the literature, and Haschke and Martz [1998] have suggested some correlations. For example, as shown in Figure 4, and given in Table I, they propose using a linear fit to three points to correlate ARFs with air velocity for temperatures greater than or equal to 500 °C. However, the three points are not all data points corresponding to tests in which only the flow velocity was varied. One point is obtained by specifying that for a zero air velocity the ARF is zero. A second point is based on the work of Chatfield in which an ARF of 7×10^{-6} is inferred by Haschke and Martz [1998] for an airflow rate of 0.45 m/s. The third point is from averaging the ARF obtained in tests M7, M8, and M9. Thus the simple linear correlations should be used with caution.

Dynamic Oxidation Experiments

All the dynamic experiments entailed burning metal fragments or droplets failing in air through a column [Carter and Stewart, 1971]. As described by these authors, the dynamic tests consisted of two types of tests:

(i) Metal droplets at about 660 °C. The specimen was suspended on an aluminum whisker, and heated in argon until it melted and fell into air.

(ii) Metal droplets at about 2000 °C. Similar to (i), but the heating was in air and continued until the metal ignited and fell.

The column dimensions and sample descriptions were given previously. Sparking was prominent in the falling droplet experiments and occurred following impact in the column. Sparking also occurred in some experiments in which the droplets were heated to 660 °C and broke apart in flight. The authors conclude that the "sparking phenomenon is the primary source of the fume fraction of the aerosol in all the dynamic experiments."

The ARF data for the dynamic tests are given in Figure 5 as circles and in the last two columns in Table II. The ARFs range from 1.5×10^{-5} to 1.0×10^{-2}, and thus the upper range of the ARF is considerably higher than the ARFs obtained for the static tests. If the tests with sparking only are considered, then from Figure 5 the ARFs range from 10^{-3} to 10^{-2}, which is higher than any of the ARFs for static tests given in Figures 2, 3, 4, and 5.

Analysis of Dynamic Oxidation Experiments

The dynamic tests allow the Pu to oxidize without contacting a solid surface that can (1) extract heat by conduction and hence reduce the temperature, (2) provide some structural support to better retain the oxide as a barrier to further oxidation, and (3) partially block the air flow. These factors apparently have a very significant effect on the ARF. Without the supporting surface, the temperature may have increased such that internal Pu vaporization with subsequent rapid expulsion caused the sparking. However, more analysis that is beyond the scope of this work is needed to quantify the influence of the supporting surface on the aerosol formation and release process.

DISCUSSION AND RECOMMENDATION

From the data we can develop a set of conditions that bound the ARF. Carter and Stewart [1971] provide such a table of conditions in which they summarize their aerosol release fraction assessments. The part of their table pertaining to plutonium is reproduced below in Table III. Notice that according to their assessment, for the 95% confidence level, the ARF varies from 1×10^{-4} when Pu is melted but remains intact, to 1×10^{-2} when droplets of burning Pu are falling in air. The suggested ARF of 1×10^{-4} by Carter and Stewart [1971] for Pu melted or ignited and burning in air without disruption into droplets is consistent with other static tests. This can be seen in Figure 4 where Carter and Stewart's suggested ARF for static oxidation is shown as a triangle. The case when droplets burn and fall in air is a dynamic oxidation process where significant fragmentation occurs, and this case is labeled "partial disruption of liquid into droplets"

in Table III. An ARF of 10^{-2} for dynamic oxidation is consistent with Figure 5 in which falling droplets have an ARF that is one to two orders of magnitude larger than that for experiments in which the Pu is static (i.e. stationary).

In Figure 5, all the static oxidation test results are given as filled-in diamonds, and the ARFs are less than ~1×10^{-4}. Therefore, if the Pu sample is stationary, then the appropriate ARF for the 95% confidence level is 1×10^{-4}. To estimate the sample size range for which this ARF applies, we note that the reported initial droplet mass ranged from 30 mg to 2.5 g. If a Pu material density of 16 g/cm^3 is used, then the droplet diameter ranged from 0.15 to 0.67 cm, respectively. We will therefore conservatively use 0.7 cm as the lower sample size limit for this ARF. (Tests with small droplets ranging in size from 0.02 to 0.04 cm falling and burning in air can result in the droplet exploding and a much higher ARF [Nelson, 1975; Nelson and Raabe, 1978; Nelson, 1980].)

Despite the experimental descriptions given by Carter and Stewart [1971], there is some confusion regarding the conditions for which an ARF of 1×10^{-2} is to be used. The Handbook references the work of Carter and Stewart [1970] for the bounding ARF of ignited-molten plutonium “disturbed by direct impact of high air velocities such as free-fall, induced high turbulence on molten surface, etc.” [DOE Handbook, page 4-2]. (The reference year in the Handbook for Cater and Stewart is 1970, but the document was also published in 1971 as given in the reference list at the end of this report.) The bounding ARF value suggested by the Handbook is based on the 95% confidence limit

recommended by Carter and Stewart and is given as 1×10^{-2}. However, such a high value for the ARF recommended by Carter and Stewart [1971] is actually for a dynamic case in which significant fragmentation occurs and not merely high turbulence on a molten surface. This is consistent with Figure 5, in which an ARF of ~1×10^{-2} bounds the falling droplet data. Therefore, it is only appropriate to use an ARF of 1×10^{-2} when the oxidizing Pu is moving freely in air, such as when droplets are sheared from the molten surface by turbulent airflow, and not merely for molten Pu exposed to turbulent airflow as given in the Handbook.

Based on all the available ARF data, and the summary analysis given by Carter and Stewart [1971], we recommend that the bounding ARF for a burning stationary mass of Pu is 2.4×10^{-3}. This is higher than the suggested value by Carter and Stewart [1971], but is recommended because it is the highest reported ARF for static Pu oxidation in air. The conditions needed to justify this value instead of an ARF of 1×10^{-2} are the following:

1. The mass of Pu remains stationary, and does not fragment into droplets less than 0.7 cm in diameter. We require that the smallest characteristic dimension of the sample be larger than 0.7 cm, because all reported tests at this size scale resulted in an ARF less than 2.4×10^{-3}. There are also no mechanical disruptions of the residue.

2. The Pu is ignited in air and not other gas mixtures. (Tests with Pu and Pu compounds in reducing and oxidizing atmospheres reported by Eidson, Yeh and Kanapilly [1988] are beyond the scope of this work.)

3. The air velocity must be less than 5.25 m/s to stay within experimentally measured conditions, even though the highest reported ARF of 2.4×10^{-3} was obtained with an air velocity of less than 0.1 m/s. (The highest reported ARF for an air velocity of 5.25 m/s was 4.7×10^{-4}.)

4. The sample temperature at all time is less than 1000 °C to be within experimentally measured conditions.

SAFETY FACTOR ESTIMATE FOR THE ACRR AND FUTURE WORK ON RECOMMENDED RELEASE ESTIMATE

Our recommended ARF for conservative safety analysis is 2.4×10^{-3} with a RF of 1. This is not a best engineering estimate, but rather a conservative safety assessment that intentionally errs on the side that would maximize the mass of respirable Pu produced by burning Pu metal in air in the ACRR. Three of the major safety factor assumptions used to obtain this recommendation are given in Table IV.

As given in Table IV, the first conservative assumption in the recommended ARF is the selection of only one experiment with the maximum ARF regardless of the experimental conditions or sample size. A more realistic best engineering estimate would use the average value of only data for samples comparable to those anticipated in the ACRR.

Such samples would be more than 100 grams, and the average ARF for such samples given for tests M7, M8 and M9 is 2.2×10^{-4}. Thus the recommended ARF is more than an order of magnitude larger than an engineering estimate. For safety analysis, however, it would be better to use the maximum measured ARF from among these tests as given by test M7. The ratio of the recommended ARF divided by that for test M7 is approximately 5. Therefore, selecting only the test for which a maximum ARF was measured as the basis for the recommended ARF of 2.4×10^{-3} has an inherent safety factor of 5.

Particle deposition on to surfaces due to diffusion, impaction, thermophoresis, electrostatic attraction, and gravitational settling are well-known removal or immobilization processes. For the recommended ARF none of these processes is considered. The safety factor introduced by neglecting these processes is difficult to estimate without more information and analysis of the experimental conditions. Therefore, listed second in Table IV, for these processes the safety factor may range from only 1 to about 5.

As given in Table I, data for samples oxidized above the ignition temperature were generally reported as completely oxidized. Thus the oxidized fraction of the original Pu mass is listed as one. However, without complete chemical analysis of the residue, it is unproven that small chunks of Pu did not break off without oxidizing. This would tend to underestimate the ARF because the ARF is based on the oxidized mass of Pu, and not the initial Pu mass. On the other hand, the descriptions of the tests indicate that at times a sample self-extinguishes by heat losses, and has to be reheated to complete the oxidation

process [Mishima, 1965]. With additional heat transfer analyses and experiments we may in the future determine that assuming complete oxidation overestimates the ARF. Therefore, listed last in Table IV, is a lower safety factor from 0.9 (which actually reduces the safety factor). This is to account for the possibility that 10% of the Pu mass that may have not been oxidized and was unaccounted for in the experiments. However, in the ACRR, if heat losses effectively extinguish oxidation, the Pu would not be intentionally reheated. Thus, a high safety factor of 10 accounts for oxidation that may be naturally self-extinguished.

If the three safety factors are independent, then the product of the lowest and highest values in the second column of Table IV give an overall safety factor in the recommended ARF of 2.4×10^{-3} that ranges from 4.5 to 250. Within these safety factors the ARF may be between $2.4 \times 10^{-3}/4.5 = 5.3 \times 10^{-4}$ to $2.4 \times 10^{-3}/250 = 9.6 \times 10^{-6}$. Thus there is significant potential that additional experiments and analyses can provide justification for using a lower recommended ARF. The analyses would involve heat transfer, aerosol transport, and reaction modeling that can be performed on today's computers, but would have not been feasible in the 1960's when the tests were performed. Furthermore, real-time aerosol instrumentation is now available that can provide detailed measurements of the release that could be correlated with time-varying sample mass and temperature. Such modeling and experimental work would provide greater safety assurance far beyond another reexamination of the limited experiments performed close to 40 years ago.

Table I. Summary of Experimental Data on Static Pu Oxidation in Air

Test	Investigator	Sample Mass (grams)	Fraction Oxidized	Aerosol Released Fraction (ARF)	Temperature (°C)	Air Flow (m/s)	Conditions
S1	Stewart, 1963	7.483	0.033	1.5×10^{-3}	123	0.05	delta alloy, 0.03 mg H_2O/liter air, 3.35 cm^2 surface area
S2	Stewart, 1963	7.344	0.054	3.2×10^{-5}	113	0.05	delta alloy, 1.5 mg H_2O/liter air, 3.30 cm^2 surface area
S3	Stewart, 1963	8.602	0.035	4.8×10^{-6}	123	0.05	delta alloy, 16 mg H_2O/liter air, 2.50 cm^2 surface area
S4	Stewart, 1963	11.021	0.57	1.4×10^{-6}	123	0.05	beta phase, 0.03 mg H_2O/liter air, 3.74 cm^2 surface area
S5	Stewart, 1963	10.802	1.0	1.1×10^{-6}	123	0.05	beta phase, 8 mg H_2O/liter air, 3.91 cm^2 surface area
S6	Stewart, 1963	7.191	0.76	3.3×10^{-6}	123	0.05	beta phase, 8 mg H_2O/liter air, 3.23 cm^2 surface area
S7	Stewart, 1963	9.397	0.21	1.3×10^{-6}	113	0.05	alpha phase, 16 mg H_2O/liter air, 3.35 cm^2 surface area
S8	Stewart, 1963	11.265	0.17	5.6×10^{-7}	123	0.05	beta phase, 16 mg H_2O/liter air, 3.78 cm^2 surface area
S9	Stewart, 1963	8.154	0.65	1.0×10^{-6}	123	0.05	beta phase, 16 mg H_2O/liter air, 3.00 cm^2 surface area

Test	Investigator	Sample Mass (grams)	Fraction Oxidized	Aerosol Released Fraction (ARF)	Temperature (°C)	Air Flow (m/s)	Conditions
S10	Stewart, 1963	12.7	1.0	1.5×10^{-5}	490 ignition 900 maximum	≤ 0.1	δ alloy billet, oxidation complete, 4.7 cm^2 surface area
S11	Stewart, 1963	10.3	1.0	1.5×10^{-5}	520 ignition 950 maximum	≤ 0.1	pure metal billet, oxidation complete, 4.7 cm^2 surface area
S12	Stewart, 1963	15.06	~0.66	2.4×10^{-3}	350 ignition 630 maximum	≤ 0.1	pure metal billet, ~2/3 oxidized, reaction not self-sustaining, 4.7 cm^2 surface area
S13	Stewart, 1963	12.2	NA	1.3×10^{-4}	515 ignition 800 maximum	≤ 0.1	δ alloy billet, reaction not self-sustaining, 4.7 cm^2 surface area
S14	Stewart, 1963	1.95	1.0	6.0×10^{-5}	180 ignition 895 maximum	≤ 0.1	δ alloy swarf, uneven heating of sample, oxidation complete, 27 cm^2 surface area
S15	Stewart, 1963	2.18	1.0	1.0×10^{-4}	110 ignition 780 maximum	≤ 0.1	δ alloy swarf, uneven heating of sample, oxidation complete, 29 cm^2 surface area
M1	Mishima, 1965	9.8944	1.0	2.8×10^{-8}	490 ignition 480 – 900 oxidation*	0.033	Pu rod 0.602 cm diameter, 1.73 cm long in, quartz combustion boat inside a furnace, 22 minute oxidation time.

Test	Investigator	Sample Mass (grams)	Fraction Oxidized	Aerosol Released Fraction (ARF)	Temperature (°C)	Air Flow (m/s)	Conditions
M2	Mishima, 1965	10.5398	1.0	3.1×10^{-7}	410 ignition 480 – 560 oxidation	0.135	Pu rod 0.594 cm diameter, 1.89 cm long in quartz combustion boat inside a furnace, 38 minute oxidation time.
M3	Mishima, 1965	10.9014	1.0	5.3×10^{-7}	495 ignition 630 – 650 oxidation	0.50	Pu rod 0.625 cm diameter, 1.85 cm long in quartz combustion boat inside a furnace, oxidation time.
M4	Mishima, 1965	11.2030	1.0	4.1×10^{-8}	495 ignition 630 – 650 oxidation	0.033	Pu rod 0.625 cm diameter, 1.89 cm long in quartz combustion boat inside a furnace, 62 minute oxidation time.
M5	Mishima, 1965	11.2796	1.0	2.6×10^{-7}	500 ignition 535 – 560 oxidation	0.033	Pu rod 0.625 cm diameter, 1.89 cm long in quartz combustion boat inside a furnace, 61 minute oxidation time.
M6	Mishima, 1965	11.3423	1.0	3.1×10^{-8}	480 ignition 520 – 590 oxidation	0.20	Pu rod 0.625 cm diameter, 1.89 cm long in quartz combustion boat inside a furnace, 64 minute oxidation time.

Test	Investigator	Sample Mass (grams)	Fraction Oxidized	Aerosol Released Fraction (ARF)	Temperature (°C)	Air Flow (m/s)	Conditions
M7	Mishima, 1966	569.8	1.0	4.7×10^{-4}	960 maximum	5.25	Alpha Pu ingot, wedge shape, 10.5 in^2 surface area. Torch ignition.
M8	Mishima, 1966	1770	1.0	1.5×10^{-4}	1000 maximum	5.25	Alpha Pu ingot, cylinder, 2.25 inch diameter, 1.5 inch long, 29.15 in^2 surface area. Torch ignition.
M9	Mishima, 1966	997	1.0	3.4×10^{-5}	930 maximum	5.25	Delta Pu ingot, hemi-cylinder, 21.73 in^2 surface area. Torch ignition.
M10	Mishima, 1966	455.5		2.9×10^{-6} **Rejected**	800 maximum	5.25	alpha Pu ingot, triangular section, 12.08 in^2 surface area. Torch ignition. **Pu covered with MgO sand after ignition. Therefore ARF not applicable.**

Test	Investigator	Sample Mass (grams)	Fraction Oxidized	Aerosol Released Fraction (ARF)	Temperature (°C)	Air Flow (m/s)	Conditions
C1	Chatfield, 1968	NA		7×10^{-6} inferred by Haschke & Martz, 1998 ARF = release rate / oxidation rate		Average flow air flow rate of 0.45 m/s.	Pu cylinder, 0.7 cm diameter and 1.0 cm long. Furnace heated.
C1	Chatfield, 1968	NA		7×10^{-6} inferred by Haschke & Martz, 1998 ARF = release rate / oxidation rate		Average flow air flow rate of 0.45 m/s.	Pu cylinder, 0.7 cm diameter and 1.0 cm long. Furnace heated.
HM1	Haschke & Martz, 1998	NA		0.6 **Rejected**	25		Personal communication with T. E. Ricketts, 1996
HM2	Haschke & Martz, 1998	NA		1.5×10^{-4} **Rejected**	500		Personal communication with T. E. Ricketts, 1996

Test	Investigator	Sample Mass (grams)	Fraction Oxidized	Aerosol Released Fraction (ARF)	Temperature (°C)	Air Flow (m/s)	Conditions
HM3	Haschke & Martz, 1998	NA		Proposes ARF = $\sim 4 \times 10^{-5}$ V (V in m/s) for temp ≥ 500 °C		Based on averaging M7, M8, and M9 for one point at 5.3 m/s, C1 at 0.45 m/s for second point, and zero ARF for zero velocity as third point.	
H1	Haschke, 1992	NA		7×10^{-4}			Author's assessment of existing data

NA = not available

*For test M1, the reported maximum oxidation temperature tabulated in the original work [Mishima, 1965] is 560 °C, however, in the same work the author twice discusses the maximum temperature for this test as 900 °C. Therefore, 900 °C is given here as the maximum oxidation temperature.

Table II. Digitized ARF Data from Carter and Stewart [1971]. The two columns on the left are static oxidation tests, and the two columns on the right are dynamic oxidation tests. The Pu sample mass ranged from 30 mg to 2.5 g in all the tests in this Table.

Cumulative % number of measurements	ARF (All Static Oxidation Results)	Cumulative % number of measurements	ARF (All 0.75 m Fall Results)
2.9	6.6×10^{-7}	4.5	1.5×10^{-5}
8.8	8.8×10^{-7}	7.3	1.9×10^{-5}
15	1.2×10^{-6}	11	2.0×10^{-5}
25	2.7×10^{-6}	15	2.9×10^{-5}
34	3.6×10^{-6}	17	4.9×10^{-5}
41	4.6×10^{-6}	20	9.9×10^{-5}
47	5.8×10^{-6}	23	1.4×10^{-4}
54	7.8×10^{-6}	27	2.1×10^{-4}
61	9.7×10^{-6}	30	7.3×10^{-4}
66	1.2×10^{-5}	34	7.3×10^{-4}
71	1.5×10^{-5}	37	8.7×10^{-4}
79	1.9×10^{-5}	40	1.1×10^{-3}
85	4.2×10^{-5}	43	1.4×10^{-3}
91	1.3×10^{-4}	46	1.6×10^{-3}
		50	1.7×10^{-3}
		53	1.8×10^{-3}
		57	2.0×10^{-3}
		59	2.3×10^{-3}
		63	2.6×10^{-3}
		66	2.8×10^{-3}
		69	3.5×10^{-3}
		72	4.1×10^{-3}
		75	4.5×10^{-3}
		79	4.5×10^{-3}
		83	4.8×10^{-3}
		87	5.0×10^{-3}
		91	5.5×10^{-3}
		93	5.9×10^{-3}
		96	8.8×10^{-3}
		99	1.0×10^{-2}

Table III. Summary of Source Fraction Involved in Aerosol Formation [Carter and Stewart, 1971].

Process	Fraction of source releases as aerosol < 10 μm unit density
Ignition and burning in air	7×10^{-5} (geometric mean) 1×10^{-4} (95% confidence limit)
Melting	7×10^{-6} (geometric mean) 1×10^{-4} (95% confidence limit)
Partial disruption of liquid into droplets (Height of fall approximately 0.75 m)	3.5×10^{-3} (geometric mean) 1×10^{-2} (95% confidence limit)
Vapor formation from droplets	About 0.5

Table IV. Conservative Safety Factor Estimates for the ACRR.

Conservative Assumption	Safety Factor	Safety Factor Basis
Use of only the one experiment with the largest ARF instead of averaging the ARFs from the experiments in the appropriate mass range.	$\frac{2.4\times10^{-3}}{4.7\times10^{-4}} = 5$	Ratio of largest ARF to largest ARF for samples larger than 100 grams is approximately 5.
All particles are uncharged and there are no aerosol removal or immobilization processes such deposition on surfaces or nonrespirable agglomerate formation.	1 - 5	Combustion aerosols may have some charge and in the tests there will be some equipment or ducts in the path of release.
Pu burns to completion without intervention and without heat sinks that effectively stop the reaction.	0.9 - 10	The tests will be actively monitored and positioned on some thermally conductive medium. There is also heat loss by convection and radiation.

Figure 1. Aerosol Particle Deposition Fractions as a Function of Aerodynamic Particle Diameter [Hinds, 1999].

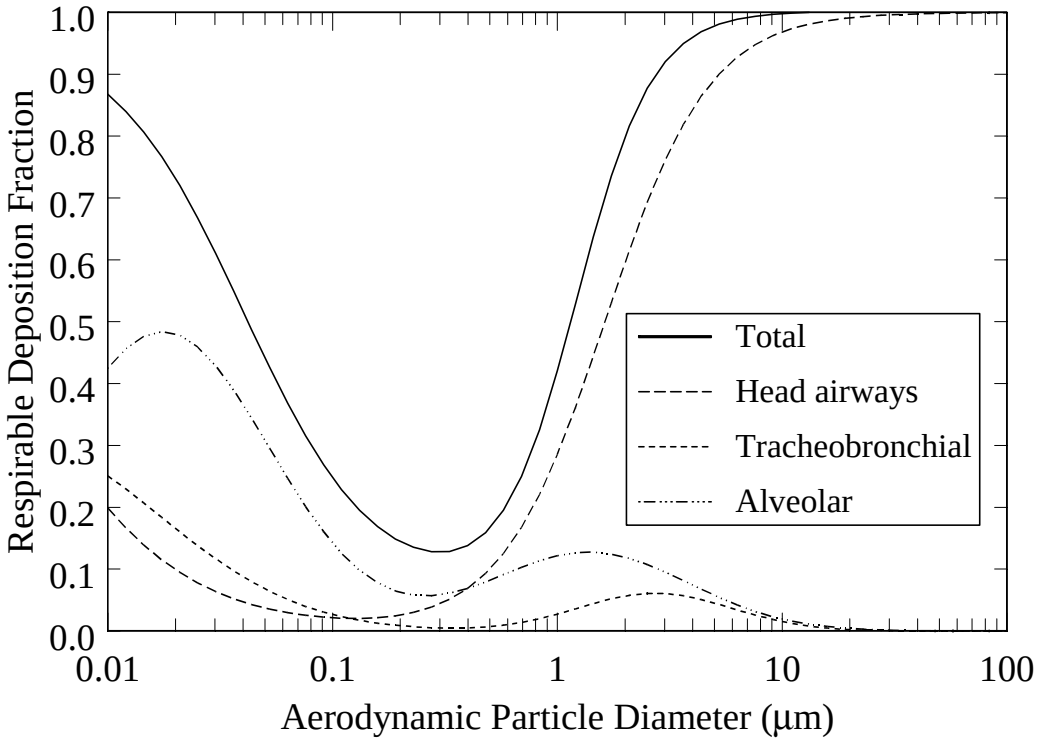


Figure 2. Experimental Data Based on Sample Mass for Static Tests.

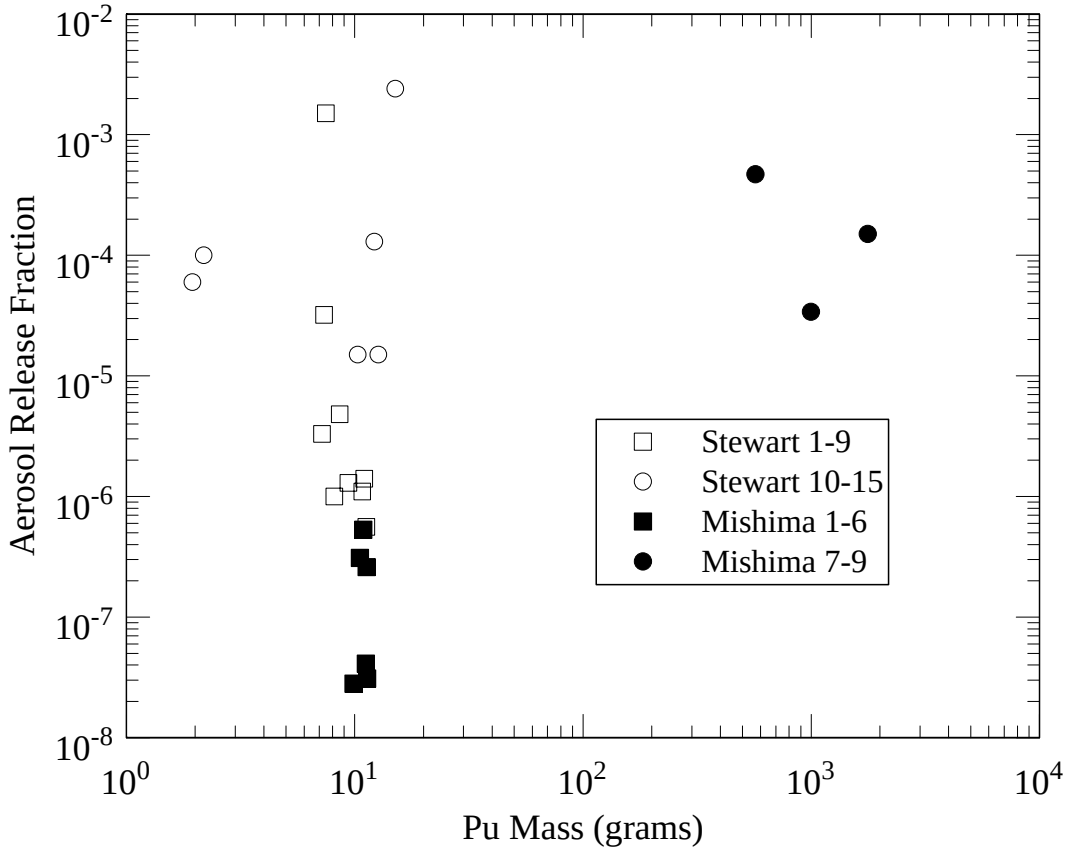


Figure 3. Experimental Data Based on Sample Temperature for Static Tests. Except for tests labeled "Stewart 1-9," the temperature varied significantly during the test and the maximum temperature is plotted.

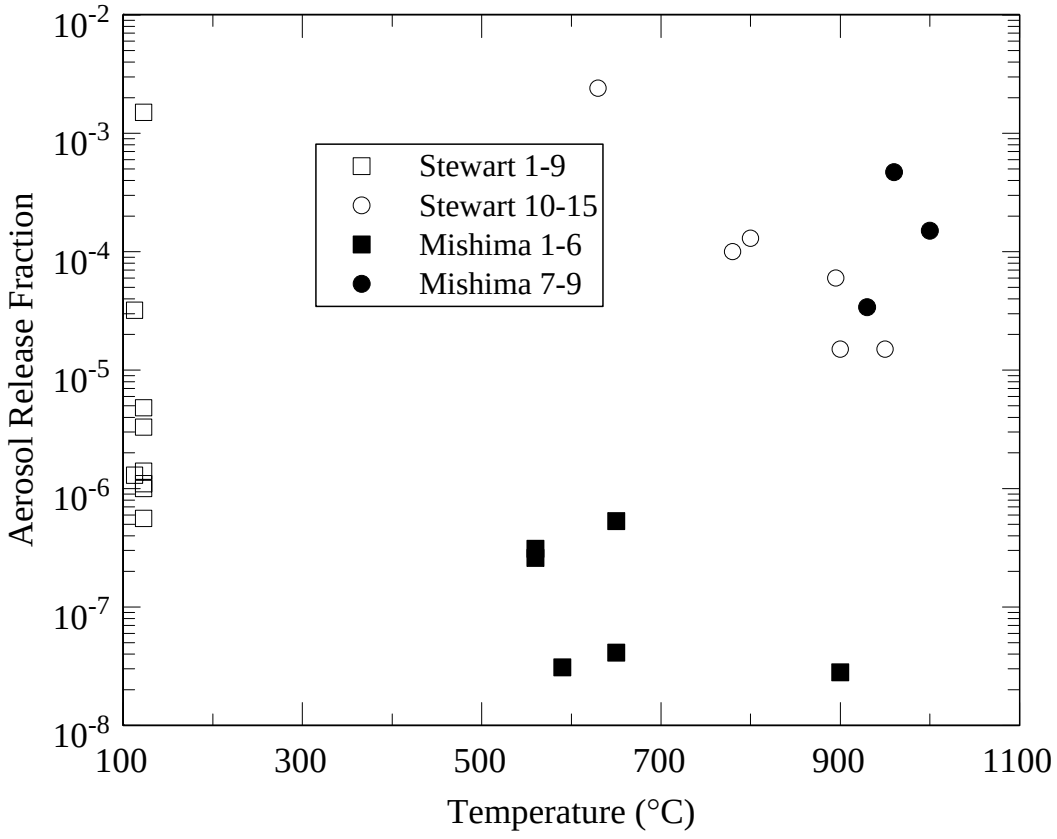


Figure 4. Experimental Data Based on Air Velocity for Static Tests.

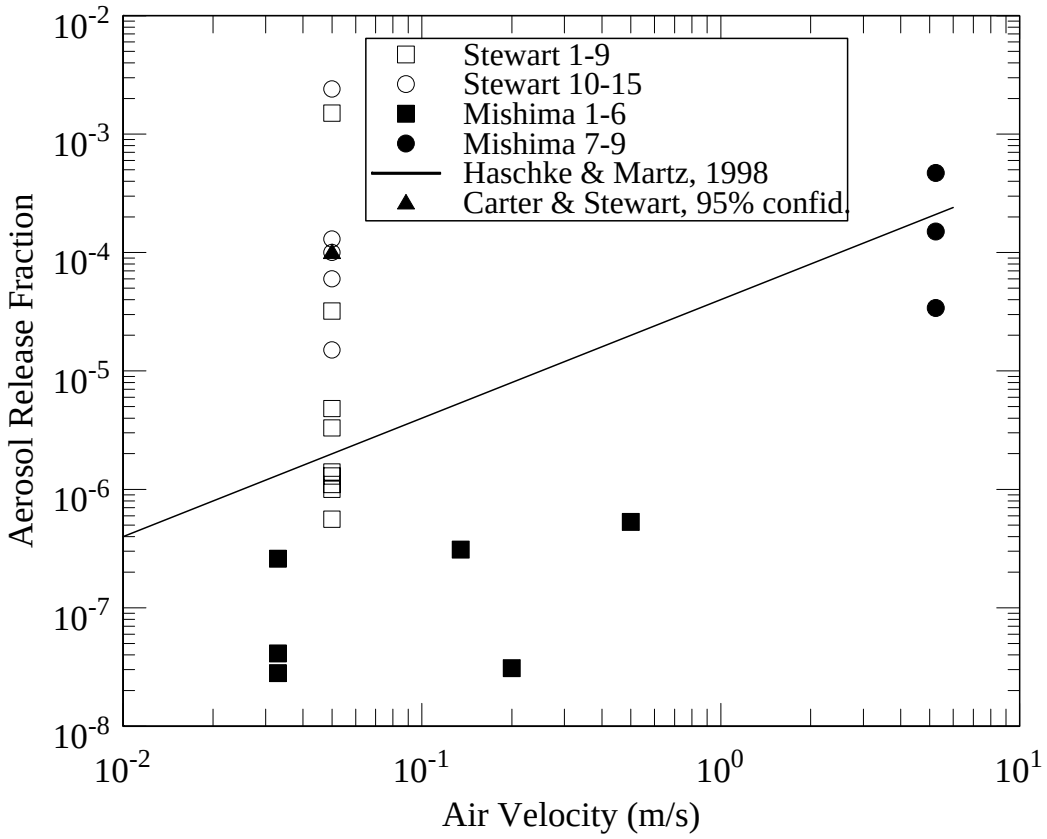
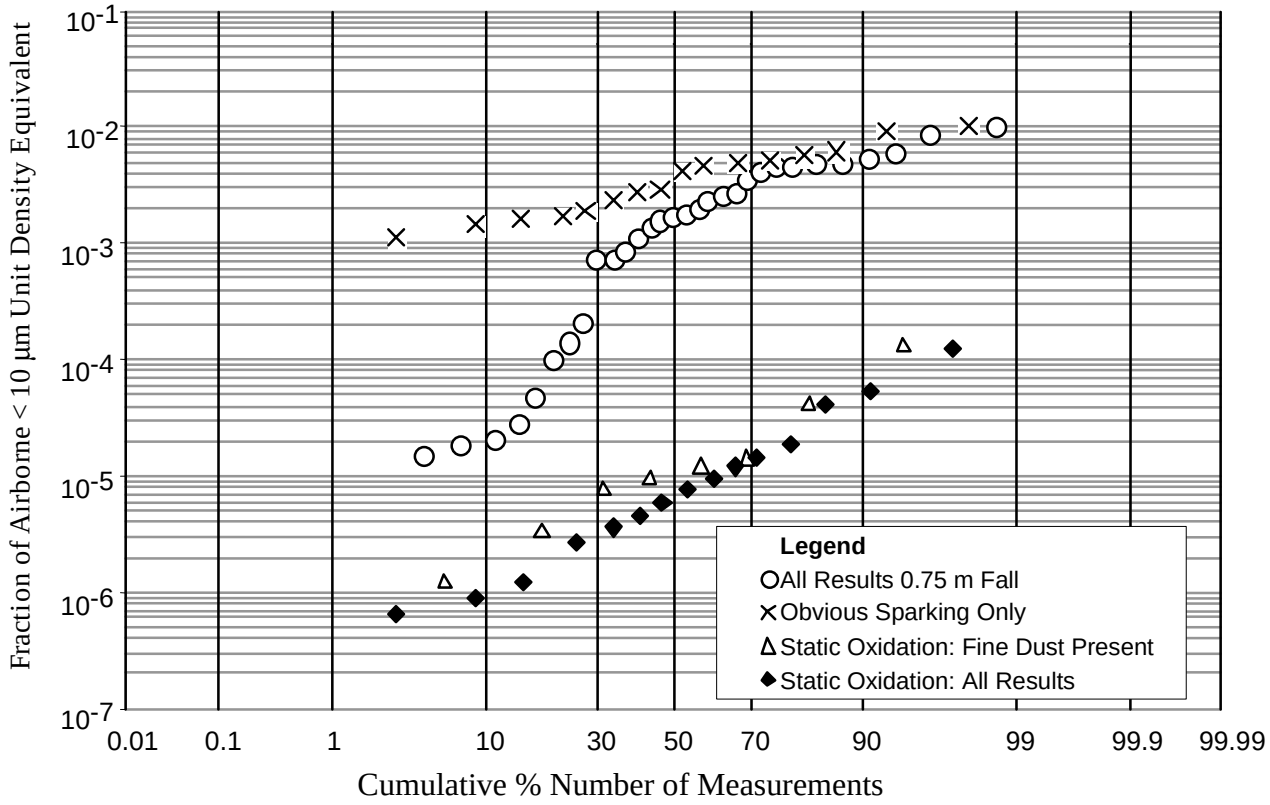


Figure 5. Redrawn Figure 13 from Carter and Stewart [1971]. The Pu sample mass ranged from 30 mg to 2.5 g (but most were in the range 40 to 200 mg), with surface area to mass ratios of 2 to 4 cm^2/g in all the tests in this Figure.



REFERENCES

Carter, R. F. and K. Stewart, "On the Oxide Fume Formed by the Combustion of Plutonium and Uranium," in W. H. Walton, Inhaled Particles III: Proceedings of an International Symposium, Unwin Brothers, 1971.

Chatfield, E. J., "The Generation of Particulate Aerosols from Plutonium Surfaces," *Atmospheric Environment*, **2**, 97-101, 1968.

DOE Handbook, "Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities: Volume I – Analysis of Experimental Data," U. S. Department of Energy, DOE-HDBK-3010-94, Washington, D. C., December 1994.

Eidson, A. F., H. C. Yeh, and G. M. Kanapilly, "Plutonium Aerosol Generation in Reducing and Oxidizing Atmospheres at High Temperatures," *Journal of Nuclear Materials*, **152**, 41-52, 1988.

Haschke, J. M., "Evolution of Source-Term Data for Plutonium Aerosolization," Los Alamos National Laboratories, Los Alamos, New Mexico, LA-12315-MS, 1992.

Haschke, J. M. and J.C. Martz, "Oxidation Kinetics of Plutonium in Air from 500 to 3500 C: Application to Source Terms for Dispersal," *Journal of Alloys and Compounds*, **266**, 81-89, 1998.

Hinds, W. C., Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles, 2nd edition, Wiley, New York, 1999.

Mishima, J., "Plutonium Release Studies I: Release From The Ignited Metal," BNWL-205, Battelle-Northwest, Richland, Washington, December 13, 1965.

Mishima, J., "Plutonium Release Studies II: Release From The Ignited, Bulk Metallic Pieces," BNWL-357, Battelle-Northwest, Richland, Washington, November 10, 1966.

Nelson, L. S., "The Combustion of Laser-Ignited Plutonium and Uranium Droplets During Free Fall Through Air," Abstract 201, Volume 75-2, The Electrochemical Society Fall Meeting, Dallas, Texas, October 5-10, 1975.

Nelson, L. S. and O. G. Raabe, "Aerosols Generated by ^{238}Pu and ^{233}U Droplets Burning in Air," *Transactions of the American Nuclear Society*, Vol. 28, Annual Meeting, June 18-22, 1978.

Nelson, L. S., "Unusually High (Oxidizer/Pu) Ratios in the Macro-Residues from Plutonium Droplet Combustion," *High Temperature Science*, **12**, 297-303, 1980.

Stewart, K., "The Particulate Material Formed by the Oxidation of Plutonium," Progress in Nuclear Energy, pp. 535-579, Pergamon Press, 1963.

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